

Contents lists available at [ScienceDirect](http://ScienceDirect.com)

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

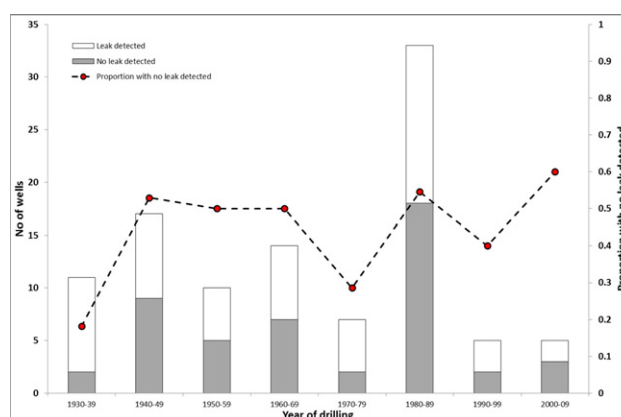
Fugitive emissions of methane from abandoned, decommissioned oil and gas wells

I.M. Boothroyd^a, S. Almond^a, S.M. Qassim^a, F. Worrall^{a,*}, R.J. Davies^b^a Centre for Research into Earth Energy Systems (CeREES), Department of Earth Sciences, Durham University, Science Labs, Durham DH1 3LE, UK^b School of Civil Engineering and Geosciences, Newcastle University, Newcastle upon Tyne, Tyne and Wear NE1 7RU, UK

HIGHLIGHTS

- This study considered the fugitive emissions from former oil and gas wells.
- 30% had CH₄ at the soil surface that was significantly larger than their respective control.
- 39% of well sites had significant lower surface soil gas CH₄ than their respective control.
- Where integrity failure occurred it appeared within a decade of well decommissioning.
- Flux of CH₄ from wells was 364 ± 677 kg CO_{2eq}/well/yr with a chance that a well was a net sink.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 20 July 2015

Received in revised form 4 December 2015

Accepted 21 December 2015

Available online 26 January 2016

Editor: Simon Pollard

Keywords:

Well integrity
Greenhouse gases
Shale gas

ABSTRACT

This study considered the fugitive emissions of methane (CH₄) from former oil and gas exploration and production wells drilled to exploit conventional hydrocarbon reservoirs onshore in the UK. This study selected from the 66% of all onshore wells in the UK which appeared to be properly decommissioned (abandoned) that came from 4 different basins and were between 8 and 79 years old. The soil gas above each well was analysed and assessed relative to a nearby control site of similar land use and soil type. The results showed that of the 102 wells considered 30% had soil gas CH₄ at the soil surface that was significantly greater than their respective control. Conversely, 39% of well sites had significant lower surface soil gas CH₄ concentrations than their respective control. We interpret elevated soil gas CH₄ concentrations to be the result of well integrity failure, but do not know the source of the gas nor the route to the surface. Where elevated CH₄ was detected it appears to have occurred within a decade of it being drilled. The flux of CH₄ from wells was 364 ± 677 kg CO_{2eq}/well/year with a 27% chance that the well would have a negative flux to the atmosphere independent of well age. This flux is low relative to the activity commonly used on decommissioned well sites (e.g. sheep grazing), however, fluxes from wells that have not been appropriately decommissioned would be expected to be higher.

© 2016 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

* Corresponding author.

E-mail address: Fred.Worrall@durham.ac.uk (F. Worrall).

1. Introduction

There are numerous environmental concerns surrounding the oil and gas industry, including the production and discharge of wastewater leading to environmental violations (Manda et al., 2014); the fugitive emission of CH₄ to the atmosphere (Caulton et al., 2014; Miller et al., 2013); and contamination of groundwater supplies (Rivard et al., 2014). It has been suggested that hydraulic fracturing, as a means of exploiting unconventional hydrocarbon resources, could be a cause of elevated CH₄ concentrations in groundwater (Osborn et al., 2011), yet it has been argued that rather than being caused by hydraulic fracturing, groundwater contamination could have been caused by other processes, one of which is well integrity failure (Davies, 2011). Well integrity refers to the zonal isolation of liquids and gases (King and King, 2013) and failure occurs when cement and/or casing barriers fail, causing a loss of zonal isolation that creates pathways for the migration of fluids, including CH₄, to groundwater, surface water and the atmosphere (Ingraffea et al., 2014). Oil and gas wells are typically constructed with multiple barriers to maintain well integrity and prevent leaks, thus well integrity failure is a consequence of complete barrier failure (King and King, 2013). Darrah et al. (2014) determined well integrity failure was the likely cause of groundwater contamination of drinking water wells overlying Marcellus and Barnett shales by CH₄ due to faulty casings and migration of hydrocarbons along the well annulus because of cement failure. Vengosh et al. (2014) also identified well integrity failure as one of the four possible risks from unconventional shale gas production to water quality and that includes well failure during and after operation and includes the risk from CH₄ leaking into groundwater. A loss of well integrity is important because it represents an uncontrolled release of fluids – whether liquid or gas – which could pose a risk to groundwater supplies and air quality. Where there is a catastrophic loss of well integrity it can cause fatalities for those close to the site. Given that natural gas is predominantly composed of CH₄, its leakage can have important consequences given its global warming potential of 24 over a 100 year timescale (Myhre et al., 2013).

There are multiple causes of a loss of well integrity. Jackson (2014) suggested that faulty casing and cementing were the cause of most leaks, with casing leaking at connections or where it has been damaged from acid corrosion. Cement can shrink (Dusseault et al., 2000) and develop cracks or channels (Jackson, 2014). Poor cement bonding between the casing and borehole has been cited as another mechanism by which wellbores lose integrity (Calosa et al., 2010; Ziemkiewicz et al., 2014) and cement bonds can deteriorate due to pressure and temperature cycling (Chilingar and Endres, 2005). Based upon the works of Celia et al. (2005); Davies et al. (2014) indicated there were seven routes by which fluid can leak from oil and gas wells: (1) between cement and surrounding rock formations; (2) between casing and surrounding cement; (3) between cement plug and casing or production tubing; (4) through cement plug; (5) through the cement between casing and rock formation; (6) across the cement outside the casing and then between the cement and the casing; and (7) along a sheared wellbore. King and King (2013) suggested that to prevent well failure pressure, temperature and corrosive environments should be properly assessed during the design phase of wells. Furthermore, Ziemkiewicz et al. (2014) argued that action to prevent integrity failure should be made appropriate to the local geology.

Reported well integrity failure rates have varied between studies. For example Erno and Schmitz (1996) found of 435 wells tested for surface casing vent leakage, 22% were leaking. Chilingar and Endres (2005) found 75% leak rates of 50 wells studied in the Santa Fe Springs oilfield which was drilled in the 1920s. Watson and Bachu (2009) analysed data from 316,439 wells drilled between 1910 and 2004 for surface casing vent flow (SCVF) through wellbore annuli and soil gas migration (GM) in Alberta and determined that 4.6% of wells suffered from surface casing vent flow or gas migration. They found that the most important cause in determining wellbore failure rates was uncemented casing.

Various estimates exist of well integrity failure in Pennsylvania. Using notices of violation from the Pennsylvania Department of Environmental Protection between January 2008 and August 2011, Considine et al. (2013) determined that of 3533 wells drilled, 2.6% experienced well integrity failure. This included four instances of blowout and venting, two instances of gas migration and 85 cement and casing violations wherein gas migration was observed. Using a similar dataset but between 2008 and March 2013, Vidic et al. (2013) found a failure rate of 3.4% from 6466 wells. Ingraffea et al. (2014) assessed 32,678 producing oil and gas wells between 2000 and 2012, finding 1.9% lost integrity during that period. Beyond the well integrity failure rate, Ingraffea et al. (2014) found that unconventional wells had six times the number of cement and casing issues compared to conventional wells. Age was also likely to increase risk of failure, with the risk increasing by 18% with each additional inspection. There were geographic factors affecting hazard risk as well, with wells drilled in north east Pennsylvania 8.5 times as likely to experience problems compared to the rest of the state. Jackson (2014) suggested that local geology and different drilling practices may have been the cause of the geographical differences in hazard risk.

Davies et al. (2014) assessed 8030 wells in Pennsylvania, indicating 6.26% had well barrier or integrity failure and 1.27% leaked to the surface. Compiling a review of all the available published sources of well barrier and integrity failure rates, Davies et al. (2014) unsurprisingly found a significant range of 1.9–75%. In the UK, of the 143 active onshore wells, only two confirmed cases of well integrity failure were found yet no monitoring of abandoned wells takes place and Davies et al. (2014) called for surveying of abandoned wells to be conducted to determine whether abandoned wells show higher rates of well integrity failure than can be determined currently. Here the term abandoned is technically correct and consistent with the literature on the subject (e.g. Davies et al., 2014). In most UK cases an abandoned well is defined as those that have been cut-off, sealed and then buried under soil and in the UK this means ~2 m of soil – in most circumstances an abandoned well might better be referred to as a decommissioned well.

Overtime it is expected that the condition of abandoned wells will deteriorate (Miyazaki, 2009) and Bishop (2013) stated that because of deterioration of well casings and cement over time, it is necessary to ensure that wells are not only properly plugged and abandoned but inspected and repaired when necessary. Post 1995, oil and gas wells in Alberta, Canada, have to undergo testing for SCVF and GM prior to final abandonment, for which wells are cut and capped (Watson and Bachu, 2009).

Little is known about the long-term integrity of abandoned wells in the UK. Of 2024 onshore wells in the UK included in the analysis of Davies et al. (2014), 65.2% were not visible as they were sealed, cut and the land reclaimed, while the remaining sites (34.8% of all known wells) retained some degree of evidence of previous drilling activity at the surface. Davies et al. (2014) suggested that surveying soils above abandoned well sites would be an important step in establishing whether there was a loss of integrity and fluid migration following well abandonment.

The aim of this study was therefore to assess whether abandoned, but properly decommissioned, wells represented an ongoing source of CH₄ to the atmosphere. The wells studied could have been exploration dry holes where no hydrocarbons were found or long-term production wells.

2. Methodology

This study selected 103 wells from across the 4 onshore UK oil and gas basins with proven oil and gas accumulations where there was more than one productive well (Fig. 1). The wells within each basin were chosen to give a range of conditions and to span the range of possible well ages (i.e. to include the oldest as well as the youngest available). One hundred and three wells were measured in this study, 102

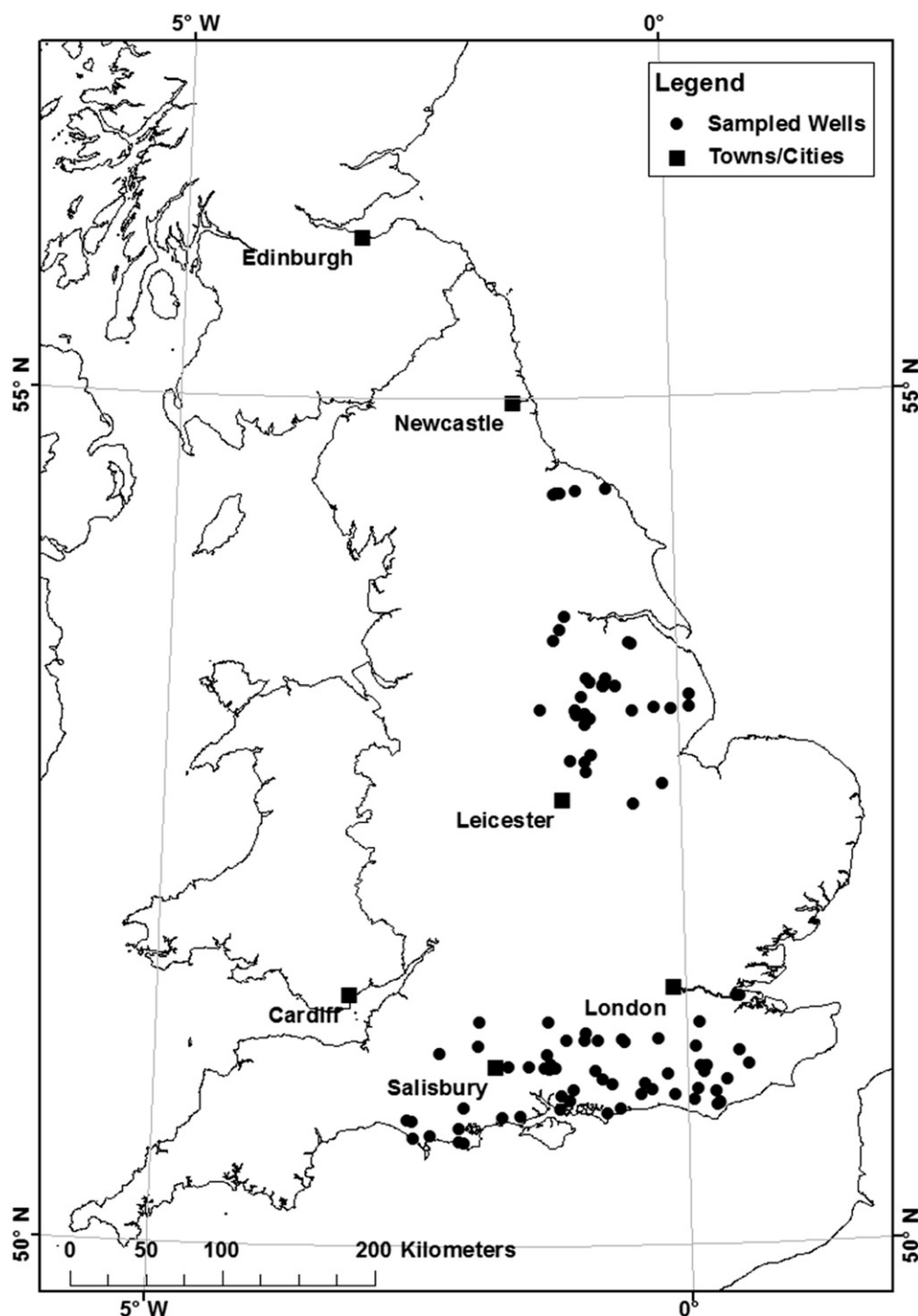


Fig. 1. Location of the well sites and basins included in this study.

of these were chosen because they had been properly decommissioned (cutoff, sealed, buried to 2 m, and vegetated including being returned to agricultural use). Of the 2024 UK onshore wells assessed by Davies et al. (2014), 65.2% were not visible at the surface, with the assumption that this was because the wells had been sealed, cut and the land reclaimed, as with the sites chosen in this study. Kang et al. (2014) considered CH₄ emissions from abandoned wells in Pennsylvania where there was a wellhead visible at the surface and as such in a UK context these would not be considered as properly abandoned and decommissioned. Similarly, Kang et al. (2015) considered plugged and unplugged oil and gas wells in Pennsylvania but this is still short of what would be

considered abandoned and decommissioned within the UK and for the purpose of this study. One well, drilled in 1917, was included in the study which had not been properly decommissioned (abandonment took place prior to the introduction of contemporary decommissioning regulations) – the well casing is visible at the surface and gases can be observed discharging through a puddle. This un-decommissioned well was included in the study as a means of assessing the CH₄ concentration within the atmosphere of the well void.

Well location information was obtained from publicly available data provided by the UK Department of Energy and Climate Change (DECC), and from the UK Onshore Geophysical Library (UKOGL) interactive

map. Discrepancies ranging from 1 m to 1358 m were noted. Where grid references differed between these two data sources, wells with a greater than 10 m discrepancy were disregarded. Given the inherent GPS error when locating well sites in the field, discrepancies of 10 m or less were considered acceptable for this study.

For each well included in the study, the agricultural field within which the well was located and the nearest field of the identical land use (most commonly this was a neighbouring field) were selected. The latter field was chosen to act as a control for the field containing the well. The control field was chosen on the basis of land use and it has been shown that CH₄ in groundwater of shale gas basins can concentrate into valleys or other hydrogeological features (e.g. Molofsky et al., 2013). In each field 7 measurements of soil gas were made equidistant along a transect of ~40 m. For the field where the well was located this transect was centred on the point directly above the known well site with 3 samples taken on either side of this centre point. The spacing of samples was matched in the control field, i.e. 7 samples equidistant apart over a distance of ~40 m but this could not be centred on any feature. It was not always possible (due to access restrictions and suitable land use) to use a separate field from the well for the control. When it was not possible to include control measurements in a separate field, the well and control shared a field. The distance between well and control in shared fields was ~40–606 m; between the well and control in separate fields the range was ~55–1405 m. Because in general control sites were chosen to be in the nearest field of the same land use as the field in which the well was located means that any difference observed between control and well fields could simply be due to the difference between any pair of fields. To test whether observed differences were simply due to differences that would be observed between any two fields, for 11 of the well sites an additional control was chosen and the soil gas measured as for the two control fields and the well field.

Soil gas concentrations of CH₄ were measured using a portable tunable diode laser (Geotechnical Instruments Ltd., Leamington Spa, UK) TDL-500 portable gas leak detector. Prior to the start of sampling on each fieldwork day, a 500 ppm \pm 5% CH₄ standard (Geotechnical Instruments Ltd., Leamington Spa, UK) was used to check the accuracy of the detector. On every occasion, the concentration detected was within the range of the standard. CH₄ was measured by connecting a telescopic rod with suction cup on the end to the TDL-500 and placing it over the ground. After 5–10 s stabilisation period, the concentration of CH₄ was recorded. No CO₂ measurements were taken within this study and it should be remembered that it is possible that decommissioned wells will also be sources of CO₂ to the atmosphere. No isotope analysis was performed and so no discrimination of the source of the measured CH₄ could be made.

At each well the air temperature, relative humidity and air pressure were measured concurrently with CH₄ concentration using a Commeter C4141 digital thermo-hygro-barometer (Comet System, s.r.o., Czech Republic).

The results from each well site were considered relative to their control field. If this study hypothesises that each control field is indicative of ambient CH₄ conditions on the day of sampling for the soil type and weather conditions then all data from each well field must be judged relative to those values. The datum for each sample from each well field was normalised to the average soil gas CH₄ concentration from the control field for each well, i.e. each of the seven soil gas measurements in each well field was transformed so that it could be interpreted as the proportion of the ambient CH₄ expected on that day for that land use and soil type.

These data were analysed by analysis of variance (ANOVA). These data were considered as a three factor experiment. The first factor was the difference between basins, this had four levels (Weald, Wessex, Malton–Pickering, and East Midlands). The second factor was site with 102 levels. Obviously it was not possible to consider each site in each basin and site was considered as a nested factor within the basin factor. The third factor was the position in field and assuming that it was

symmetric about the sampling location above the well head. Again the position in field was considered as a nested factor and had four levels (labelled as 3, 2, 1 and 0, where 0 is the location over the well and 3 is the location furthest from the well). The significance of individual factors and interactions between factors were considered and the magnitude of the effects calculated using generalised ω^2 (Olejnik and Algina, 2003). The underlying assumption of ANOVA is that all variance is random and that factors and interactions have to be proved (at given probability) to be significant. This means that the approach is conservative and also that the unexplained variance can be measured relative to the importance of the factors and interactions. Post-hoc testing of the results between factor levels, using Tukey's pairwise comparisons, was conducted to assess where significant differences lay between factor levels. Prior to analysis the data were Box–Cox transformed to remove outliers; the Anderson–Darling test was used to confirm normality; and the Levene test was used to assess homogeneity of variance. If either of these tests were failed then the data were log-transformed and re-tested. Secondly, to avoid type I errors all probability values were given even if significance were assessed at the 95% level. Thirdly, a power analysis was used to assess the minimum effect size that could be detected within this design. The study was fully factorial with respect to each of 3 factors, 1 centre point was assumed; the standard deviation was estimated as the square root of the mean square difference; and the required experimental power was set at 80%. Results are expressed as least squares means as these are better estimates of the mean for that factor level having taken account of the other factors, interactions and covariates that were included in the analysis. All statistical modelling was performed using Minitab v16 (Minitab Ltd, Coventry, UK). The ANOVA was then repeated including covariates. The covariates included the meteorological conditions measured at the time of sampling on each well site (the air temperature, relative humidity and air pressure) and the age of the well. It should be noted that when covariates were included in the analysis then site factor was not included as there would be exact co-linearity between the site factor and the well age. The covariates were tested for normality as above and log-transformed as necessary.

In a second ANOVA, the 11 sites with two control fields (henceforward referred to as control fields (a) and (b)) were considered. In this ANOVA, an additional factor was included; this factor henceforward referred to as field type, had three levels: control (a), control field (b), and the well field. All CH₄ measurements recorded for these 11 sites were judged relative to the first control field (control field (a)) at each site and then all other factors were considered as before. If there was a significant effect that could be simply ascribed to differences between fields as opposed to a difference between a well field and a control field then we would expect a significant difference between control fields (a) and (b). If this study's assumption that any observed difference between well and control field can be ascribed to the well then we would expect no significant difference between control fields (a) and (b) and the well field.

2.1. Flux modelling

Given the soil gas measurements the flux of CH₄ from the soil surface was considered using Fick's first law:

$$J = D \nabla \phi \quad (1)$$

where: J = the diffusive flux (mg CH₄/m²/s); D = diffusion coefficient (m²/s); and ϕ = the concentration of CH₄ in soil (mg CH₄/m³). Eq. (1) was solved assuming that the flux was at steady state over time in 2-dimensions using an explicit finite difference method with Δx and $\Delta y = 0.1$ m. The boundary conditions were chosen such that ϕ was at the ambient CH₄ concentration as measured for the control field. The decommissioned well was located at the centre of the base of the grid and the central grid cell was given a concentration equivalent to that

in the well at a depth of 2 m as required for UK decommissioning. Firstly, the model was developed fitting the observed values of ϕ assuming observed values for equivalent to ϕ at 10 cm depth; the concentration in the well was taken as the maximum value observed in the field measurements; and using D as a fitting parameter. Secondly, the value of D was set based upon the approach proposed by Ridgewell et al. (1999):

$$D_{\text{soil}} = 0.196(1 + 0.0055T_{\text{soil}})f_{\text{air}}^4 \left(\frac{f_{\text{air}}}{f} \right)^{1.5 + \frac{3}{b}} \quad (2)$$

$$b = 15.9f_{\text{clay}} + 2.91 \quad (3)$$

where: T_{soil} = ambient temperature ($^{\circ}\text{C}$); f = fractional total porosity; f_{air} = fractional air-filled porosity; f_{clay} = fraction of clay-sized particles in the soil. The value of T_{soil} was taken as the average median daily temperature from the CET = 9.1°C (Parker et al., 1992). The value of f_{clay} was 0.3 based on the soil being a loam (Avery, 1980). For the UK a typical loam soil will have a total porosity of 0.52. In this configuration the concentration in the well was taken as the fitting parameter.

3. Results

In total 1529 CH_4 measurements were made with 804 measurements on control fields and 725 in well fields. On the control fields the median value was $1.4 \text{ mg CH}_4/\text{l}$ with an interquartile range from 1.2 to $1.7 \text{ mg CH}_4/\text{l}$ while for the well fields the median value was 1.4 with an interquartile range between 1.2 and $1.7 \text{ mg CH}_4/\text{l}$, i.e. exactly the same as for the controls. When relative concentrations were considered then the median was 0.97 with an interquartile range of 0.78 to 1.17. However, this is still misleading and the median value for the location directly above the well was 1 with an interquartile range between 0.78 and 1.20, and dropping to a median of 0.95 (0.78 to 1.16) for 20 m from the well head. Of the 102 wells visited 50 had relative concentrations above their well head of greater than 1.00.

The Box–Cox transformation removed only one well out of the entire dataset; Hardstoft No.1 at Tibshelf which is the oldest recorded oil or gas well in the UK and therefore was not decommissioned under the same regulatory regime as other wells in the study. The Hardstoft No.1 well was directly venting to the surface through a puddle of water via

ebullition. These bubbles were measured at concentrations of $50 \text{ mg CH}_4/\text{l}$ – a relative concentration of 36.5. This data was not included in the ANOVA but was used as an estimate of the source term in the diffusion modelling. The power analysis suggests that at 80% probability this experimental design could find a maximum difference of 0.19 between well sites below the maximum difference actually observed of 1.59.

The results of ANOVA show that all factors were significant but no interactions were found to be significant. The basin factor explained 6.5% of the variance in the original dataset. Post-hoc testing of the basin data showed that the Weald basin was significantly higher than both the East Midlands and Wessex basin; and the Malton–Pickering basin was also significantly higher than the Wessex basin (Fig. 2). The Weald sites were on average 13% higher than ambient while the Wessex sites were 8% lower than ambient. The distance factor explained only 0.5% of the variance in the original dataset and post-hoc testing showed that it was only the position over the well that was significantly higher than all other sampling positions, with the position over the well being 7% higher than ambient and the least squares mean of all other positions being within 1% of ambient.

The most important factor was the difference between well sites which explained 73% of the variance in the original dataset (Fig. 3). For 31 of the wells the results were significantly greater than ambient with the maximum observed being 147% higher than ambient. Thirty nine wells were significantly lower than ambient, with the lowest 63% below ambient.

The ANOVA based upon three factors explained 81% of the variance in the original dataset with an error term explaining 19% of the original variance – a mean effect of 0.026 in the relative CH_4 measurement. This error term represents the unexplained variance and not only represents measurement error but also sampling error or any factors and interactions that were not, or could not be, included in the model.

When the ANOVA was repeated including covariates then none of the meteorological covariates were found to be significant. Because the sampling design of this study could not be cross-classified with respect to sites the covariates had therefore been included as a method of comparing between days of sampling and it had been hypothesised that the difference between sites and basins was really due to differences in the weather on individual sampling days. The lack of significant meteorological covariates means that the differences between basins

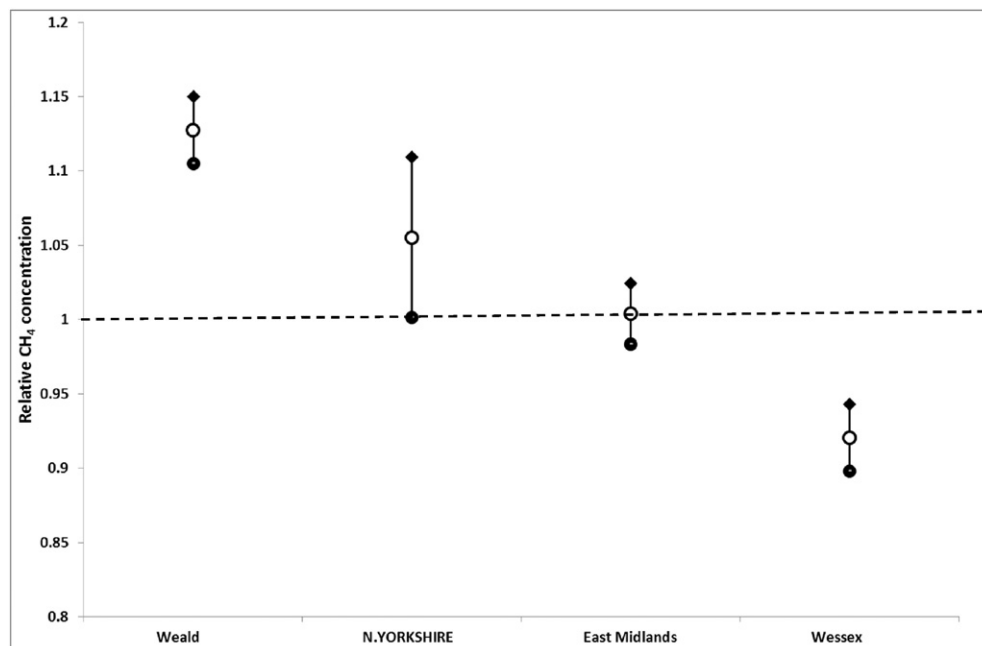


Fig. 2. The main effects plots of the basin factor. Results are given as the least squares mean and the standard error on that mean.

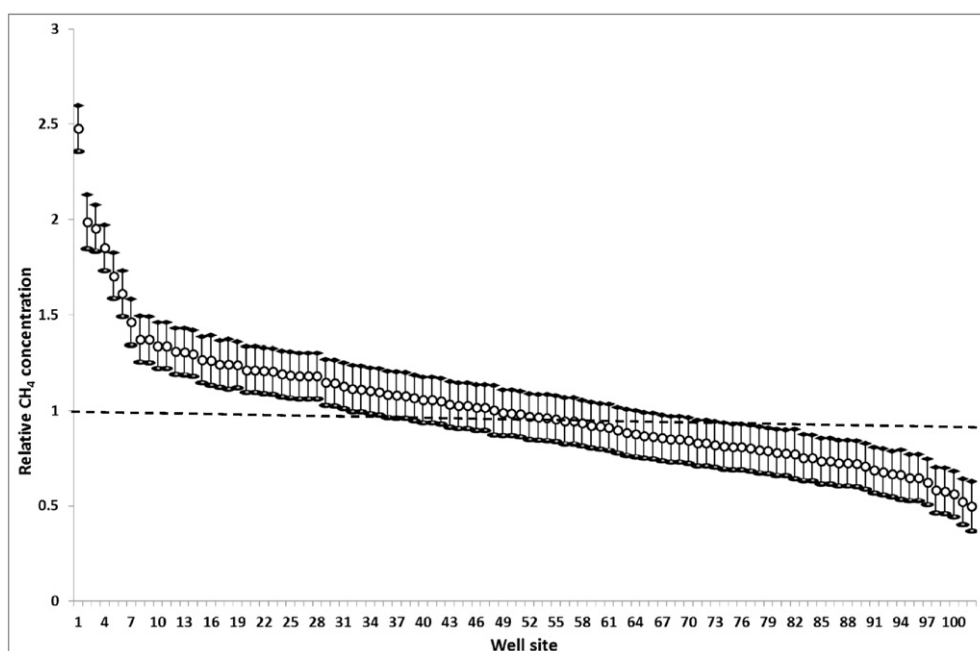


Fig. 3. The main effects plots of the well site factor. Results are given as the least squares mean and the standard error on that mean.

and sites does reflect processes other than meteorological conditions on the day of the sampling and observed differences cannot simply be ascribed to the different days upon which measurements were done. The well age was not a significant covariate.

When the 11 sites with a second control field were considered then the field type factor was significant (at $P > 95\%$) and was the second most important factor after the difference between sites. Post-hoc testing shows that the significant difference in the field type factor was not between control field (a) and control field (b) but between both control

field (a) and (b) and the well field. Thus the assumption that observed differences in the larger, 102 well dataset are due to the well field is upheld by this analysis and the hypothesis that differences between control and well fields is just a difference between fields can be rejected.

When considered as a proportion then the failure rate can be plotted against time (Fig. 4). As was discovered when well age was included as a covariate in the ANOVA above, the failure shows no significant trend over time. Failure rate was highest for the oldest wells within the survey (82% for those wells drilled between 1930 and 1939) but the failure rate

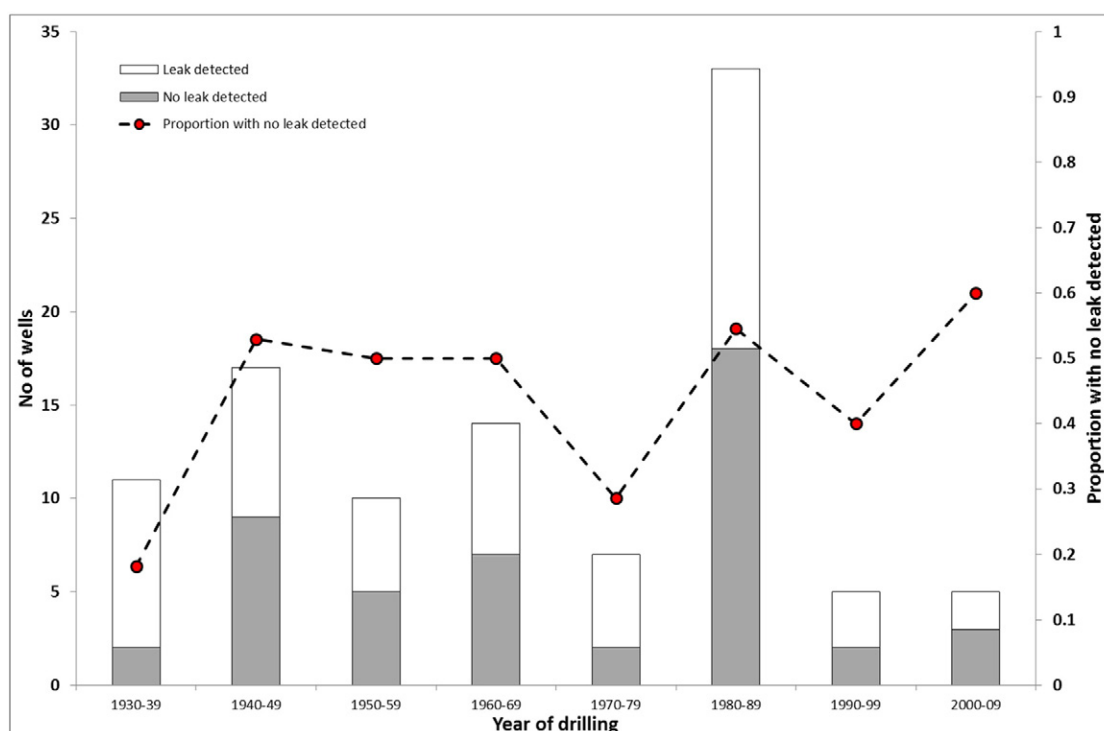


Fig. 4. The relative CH_4 concentration for the sample directly above the well head for each site compared to the age since abandonment.

was 47% for those wells drilled between 1940 and 1949 and 40% for the most recently drilled wells in the dataset. This implies that when well integrity is a problem it occurs early on in the decommissioned life of a well, although it should be noted that the study only had access to the drilling date of each well and not its actual date of decommissioning.

3.1. Diffusive modelling

Given Eqs. (2) and (3) the value of $D_{\text{soil}} = 0.086 \text{ cm}^2/\text{s}$ and given a concentration of CH_4 at depth equivalent to that found for Hardstoft gives a value of CH_4 for the value at the source, i.e. in the well the concentration of CH_4 was $50 \text{ mg CH}_4/\text{l}$, and an ambient atmospheric concentration CH_4 of $1.5 \text{ mg CH}_4/\text{l}$. These values would give a surface soil concentration of $1.53 \text{ mg CH}_4/\text{l}$ – a relative concentration of 1.02. In this configuration the plume from the well is no more than 2 m wide and would represent a flux of $49 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. However, the relative concentration is lower than observed and at the mean relative concentration of 1.067 then the D_{soil} has to be increased to 0.215 with a source concentration of $50 \text{ mg CH}_4/\text{l}$, this gives a flux of $362 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. Alternatively, leaving relative concentration as 1.067 and $D_{\text{soil}} = 0.086 \text{ cm}^2/\text{s}$ then this requires a source concentration = $159 \text{ mg CH}_4/\text{l}$ leading to a flux of $146 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. The largest relative concentration observed across all wells was 2.47 which at a $D_{\text{soil}} = 0.086 \text{ cm}^2/\text{s}$ gives an initial concentration – $3500 \text{ mg CH}_4/\text{l}$ (0.43%) and a flux of $3256 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. The lowest relative concentration observed across all wells was 0.65 which at a $D_{\text{soil}} = 0.086$ gives an initial concentration = $0 \text{ mg CH}_4/\text{l}$ and gives a sink of $-563 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. Calculating the flux for the least squares mean relative concentrations for each well shows that the distribution across all wells is normal with a mean of $364 \pm 677 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$ where the uncertainty is given as the standard deviation in the mean – given the distribution there is a 28% chance that any well would be a net sink of CH_4 . Note that this latter value is across all wells across all ages.

4. Discussion

There are a number of reasons why a well site may show soil gas concentrations lower than the local ambient value and CH_4 oxidation is a common phenomenon in soils (eg. Curry, 2009). Firstly, well sites will not just consist of a well but would have also had associated works and thus decommissioned sites may have replaced soils but these soils may be distinct from those that have never had to develop over a former well site. On a decommissioned well site there may be a lack of deep soil, meaning soil gas concentrations lower than ambient values represent a lack of production. Poor soil development over the decommissioned site may lead to soils with low organic matter and poor soil structure and therefore lacking sites of reduction. CH_4 oxidation is commonly observed in UK mineral soils: Levy et al. (2012) conducted a large meta-analysis of CH_4 flux measurements from 21 sites across the UK including 8 sites based on mineral soils with an average of 238 measurements per site and when rescaled for 1 year the results varied from a CH_4 sink of $0.5 \text{ g CH}_4/\text{m}^2/\text{year}$ to a net source of $1.4 \text{ g CH}_4/\text{m}^2/\text{year}$. Ball et al. (2002) found net CH_4 sinks of between 0.025 and $0.27 \text{ g CH}_4/\text{m}^2/\text{year}$ for mineral soils under grass and arable management. Alternatively, where the well site gives values below that expected this could be classed as an avoided loss, i.e. the soil is not extracting CH_4 from the atmosphere but has perhaps evolved to remove CH_4 from a leaking well.

Many of the decommissioned sites visited in this study had been converted to grassland for agricultural production. There is currently no emissions factor for CH_4 from grassland or arable soils in the UK – the emissions are assumed to be from the housing of livestock, storage of manures, manure application, and fertiliser but not from the soil and vegetation itself (Sneath et al., 1997; Chadwick et al., 1999). For dairy breeding herd the CH_4 emissions factor (enteric and manure

sources) is $128 \text{ kg CH}_4/\text{head}/\text{year}$ ($2944 \text{ kg CO}_{2\text{eq}}/\text{head}/\text{year}$) while for a breeding ewe the value is $8.9 \text{ kg CH}_4/\text{head}/\text{year}$ ($205 \text{ kg CO}_{2\text{eq}}/\text{head}/\text{year}$). For lowland agriculture in the UK typical grazing rates are 3 cows or 21 sheep per hectare. If the decommissioning of the wells considered in this study had brought 1 ha of land back into livestock production then the increase in herd size would considerably dominate over the emissions from the decommissioned well in that field.

Emission factors for fugitive emission from natural gas production are normally expressed relative to energy production, e.g. $\text{g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$. MacKay and Stone (2013) summarised the likely impact that shale gas production would have upon carbon emissions and compared it to other sectors in the energy industry. The carbon footprint of shale gas (under a 90% capture and flare scenario of CH_4 released during completion) was expected to be $200\text{--}253 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$, which was comparable to conventional natural gas ($199\text{--}207 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$) and lower than liquid natural gas ($233\text{--}270 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$). These figures included $190 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$ from combustion of the gas. The contribution of preproduction and processing varied, depending upon whether the results of Howarth et al. (2011) were included, because of the influence of results predicted from well completion of Haynesville shale gas. The predicted emission of $102,000 \text{ t CO}_{2\text{eq}}/\text{well}$ was well beyond the next nearest amount, also for Haynesville, of $18,000 \text{ t CO}_{2\text{eq}}/\text{well}$ (O'Sullivan and Paltsev, 2012). Thus, under a 90% capture and flare scenario, the mean emissions intensity under a central productivity value of 85 million m^3 varied between 14 and $19 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$, with a range of $10\text{--}88 \text{ g CO}_{2\text{eq}}/\text{kWh}_{(\text{th})}$ under high and low productivity. Jiang et al. (2011) modelled lifecycle emissions from shale gas wells which ranged from $65.3\text{--}74.4 \text{ g CO}_{2\text{eq}}/\text{MJ}$, which equates to $236\text{--}269 \text{ g CO}_{2\text{eq}}/\text{kWh}$. Of all the scenarios of productivity and well lifetime, the average (base case) contribution of preproduction emissions in Jiang et al. (2011) was $1.8 \text{ g CO}_{2\text{eq}}/\text{MJ}$, or $6 \text{ g CO}_{2\text{eq}}/\text{kWh}$. The largest contributor to the lifecycle emissions was combustion of the gas, at $50 \text{ g CO}_{2\text{eq}}/\text{MJ}$, or $181 \text{ g CO}_{2\text{eq}}/\text{kWh}$ which is in agreement with MacKay and Stone (2013).

The contribution of preproduction emissions varied in MacKay and Stone's (2013) analysis of emissions from shale gas, depending upon whether the results of Howarth et al. (2011) were included, because of the influence of results predicted from well completion of Haynesville shale gas. The predicted well completion emission of $102,000 \text{ t CO}_{2\text{eq}}/\text{well}$ was well beyond the range of emissions from the other sources used. The other well completion emissions included $9100 \text{ t CO}_{2\text{eq}}/\text{well}$ for Marcellus shale (Jiang et al., 2011), 5600 for Barnett shale (Howarth et al., 2011), and a range of $4100\text{--}18,000 \text{ t CO}_{2\text{eq}}/\text{well}$ across numerous sites, with the latter from Haynesville (O'Sullivan and Paltsev, 2012). Emissions from well completion, let alone lifecycle emissions, are much higher than those associated with well leakage of $364 \pm 677 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$, following abandonment.

It should be remembered that this study chose to study wells which had, a priori, been decommissioned in line with current best practice recommendations in the UK. Davies et al. (2014) has noted that some onshore wells in the UK showed clear visual evidence of not having been appropriately decommissioned – in most cases this meant that the well pad was still visible from aerial photographs. This study must assume that fluxes from such legacy sites will be greater than those considered here, and indeed the study deliberately chose to include one well that was still open at the surface and given its values the estimated flux would be $8604 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$. Kang et al. (2014) in their study of abandoned oil and gas wells in Pennsylvania found an equivalent value of $11.3 \text{ kg CO}_{2\text{eq}}/\text{well}/\text{year}$ but given the number of abandoned wells within Pennsylvania the flux from abandoned wells (although not decommissioned ones) would represent between 4 and 7% of total emissions from the state. It should also be remembered that this study considered the vertical emissions from well integrity failure but not the potential for diffuse leakage into the surrounding groundwater and enhanced release over a broad area. Kang et al. (2015) have measured effective permeabilities which combine the pathways from

within and around the wellbore of plugged and unplugged, abandoned oil and gas wells (not decommissioned in the UK sense) and found permeabilities between 1.2×10^{-6} and 120 mm/day.

It was not possible to determine the specific details of the well design and the decommissioning process, such as the depth that cement plugs were set. In the supplementary material, Table S1 lists petroleum, gas and borehole legislation onshore from the Petroleum (Consolidation) Act 1928 to the Environmental Permitting (England and Wales) (Amendment) Regulations from 2015. Not all legislation is relevant to well decommissioning or well integrity, though where such legislation does incorporate decommissioning, details typically refer to industry best practice for abandonment or standards set out by the Minister or Health and Safety Executive rather than a technical specification. Furthermore, it can also be difficult to obtain information on individual wells given discrepancies in records between the UK Department of Energy and Climate Change and the UK Onshore Geophysical Library and as highlighted in the methodology.

5. Conclusions

The study has detected elevated concentrations of soil gas methane above decommissioned (abandoned) oil and gas wells. The study showed that for 31 of the 102 wells (30%) the soil gas CH₄ was significantly ($P > 95\%$) higher than that for their respective control sites with the maximum observed being 147% greater than the control. In contrast, 39 out of 102 wells showed significantly lower soil gas CH₄ than their respective controls indicating that soils on some decommissioned sites would act as a net CH₄ sink. The modelled fluxes from the well sites suggest a mean fugitive emission of 364 ± 677 kg CO_{2eq}/well/year where the uncertainty is given as the standard deviation in the mean – given the distribution there is a 27% chance that any well would be a net sink of CH₄. The estimated fugitive emissions from decommissioned wells are less than that for the agricultural activities that would take place on the reconstituted land. The relative CH₄ concentration above wells did not significantly increase with the age of the well since drilling and 40% of the most recent wells surveyed showed leaks implying that leaks develop early in the post-production life of a decommissioned well. For a well that had not been decommissioned the CH₄ flux was 8604 kg CO_{2eq}/well/year.

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2015.12.096>.

Acknowledgements

This research was carried out as part of the ReFINE (Researching Fracking in Europe) consortium led by Durham University and Newcastle University and has been funded by the Natural Environment Research Council (UK), Shell, Chevron, GDF Suez, Ineos, Centrica and the Environment Agency. We are grateful to ethics committees in Durham University's Faculty of Science and Newcastle University's Faculty of Science, Agriculture and Engineering for their time in providing advice on research ethics. We thank the ReFINE Independent Science Board (<http://www.refine.org.uk/how-we-work/independent-scienceboard>) for spending time prioritising the research projects undertaken by ReFINE. The authors would like to thank Tony Almond at the Health and Safety Executive for compiling all relevant legislation for use in this paper. We are grateful to all the land owners who allowed access to their fields, enabling this research to be undertaken. We thank two anonymous referees for assistance in editing and reviewing the manuscript.

References

Avery, B.W., 1980. Soil Classification for England and Wales (Higher Categories). Soil Survey Technical Monograph No. 14 (Harpenden).
 Ball, B.C., McTaggart, I.P., Watson, C.A., 2002. Influence of organic ley-arable management and afforestation in sandy loam to clay loam soils on fluxes of N₂O and CH₄ in Scotland. *Agric. Ecosyst. Environ.* 90 (3), 305–317.

Bishop, R.E., 2013. Historical analysis of oil and gas well plugging in New York: is the regulatory system working? *New Solutions* 23 (1), 103–116.
 Calosa, W.J., Sadarta, B., Ronaldi, R., 2010. Well Integrity Issues in Malacca Strait Contract Area Society of Petroleum Engineers.
 Caulton, D.R., Shepson, P.B., Santoro, R.L., Sparks, J.P., Howarth, R.W., Ingraffea, A.R., Cambaliza, M.O.L., Sweeney, C., Karion, A., Davis, K.J., Stirm, B.H., Montzka, S.A., Miller, B.R., 2014. Toward a better understanding and quantification of methane emissions from shale gas development. *Proc. Natl. Acad. Sci. U. S. A.* 111 (17), 6237–6242.
 Celia, M.A., Bachu, S., Nordbotten, J.M., Kavetski, D., Gasda, S.E., 2005. Modeling critical leakage pathways in a risk assessment framework: representation of abandoned wells. Fourth Annual Conference on Carbon Capture and Sequestration DOE/NETL.
 Chadwick, D.R., Sneath, R.W., Phillips, V.R., Pain, B.F., 1999. A UK inventory of nitrous oxide emissions from farmed livestock. *Atmos. Environ.* 33, 3345–3354.
 Chilingar, G.V., Endres, B., 2005. Environmental hazards posed by the Los Angeles Basin urban oilfields: an historical perspective of lessons learned. *Environ. Geol.* 47 (2), 302–317.
 Considine, T.J., Watson, R.W., Considine, N.B., Martin, J.P., 2013. Environmental regulation and compliance of Marcellus shale gas drilling. *Environ. Geosci.* 20 (1), 1–16.
 Curry, C.L., 2009. The consumption of atmospheric methane by soil in a simulated future climate. *Biogeosciences* 6 (11), 2355–2367.
 Darrah, T.H., Vengosh, A., Jackson, R.B., Warner, N.R., Poreda, R.J., 2014. Noble gases identify the mechanisms of fugitive gas contamination in drinking-water wells overlying the Marcellus and Barnett Shales. *Proc. Natl. Acad. Sci. U. S. A.* 111 (39), 14076–14081.
 Davies, R.J., 2011. Methane contamination of drinking water caused by hydraulic fracturing remains unproven. *Proc. Natl. Acad. Sci. U. S. A.* 108 (43), E871.
 Davies, R.J., Almond, S., Ward, R.S., Jackson, R.B., Adams, C., Worral, F., Herringshaw, L.G., Gluyas, J.G., Whitehead, M.A., 2014. Oil and gas wells and their integrity: Implications for shale and unconventional resource exploitation. *Mar. Pet. Geol.* 56, 239–254.
 Dusseault, M.B., Gray, M.N., Nawrocki, A., 2000. Why Oilwells Leak: Cement Behavior and Long-Term Consequences. Society of Petroleum Engineers.
 Erno, B., Schmitz, R., 1996. Measurements of soil gas migration around oil and gas wells in the Lloydminster area. *J. Can. Pet. Technol.* 35 (7), 37–46.
 Howarth, R.W., Santoro, R., Ingraffea, A., 2011. Methane and the greenhouse-gas footprint of natural gas from shale formations. *Clim. Chang.* 106 (4), 679–690.
 Ingraffea, A.R., Wells, M.T., Santoro, R.L., Shonkoff, S.B.C., 2014. Assessment and risk analysis of casing and cement impairment in oil and gas wells in Pennsylvania, 2000–2012. *Proc. Natl. Acad. Sci. U. S. A.* 111 (30), 10955–10960.
 Jackson, R.B., 2014. The integrity of oil and gas wells. *Proc. Natl. Acad. Sci. U. S. A.* 111 (30), 10902–10903.
 Jiang, M., Griffin, W.M., Hendrickson, C., Jaramillo, P., VanBriesen, J., Venkatesh, A., 2011. Life cycle greenhouse gas emissions of Marcellus shale gas. *Environ. Res. Lett.* 6 (3), 9.
 Kang, M., Kanno, C.M., Reid, M.C., Zhang, X., Mauzerall, D.L., Cella, M.A., Chen, Y., Onstott, T.C., 2014. Direct measurements of methane emissions from abandoned oil and gas wells in Pennsylvania. *Proc. Natl. Acad. Sci. U. S. A.* 111 (51), 18173–18177.
 Kang, M., Baik, E., Miller, A.R., Baudilla, K.W., Celia, M.A., 2015. Effective permeabilities of abandoned oil and gas wells: analysis of data from Pennsylvania. *Environ. Sci. Technol.* 49, 4757–4764.
 King, G.E., King, D.E., 2013. Environmental risk arising from well-construction failure-differences between barrier and well failure, and estimates of failure frequency across common well types, locations, and well age. *SPE Prod. Oper.* 28 (4), 323–344.
 Levy, P.E., Burden, A., Cooper, M.D.A., Dinsmore, K.J., Drewer, J., Evans, C., Fowler, D., Gaiawyn, J., Gray, A., Jones, S.K., Jones, T., McNamara, N.P., Mills, R., Ostle, N., Sheppard, L.J., Skiba, U., Sowerby, A., Ward, S.E., Zielinski, P., 2012. Methane emissions from soils: synthesis and analysis of a large UK data set. *Glob. Chang. Biol.* 18 (5), 1657–1669.
 MacKay, D.J.C., Stone, T.J., 2013. Potential Greenhouse Gas Emissions Associated With Shale Gas Extraction and Use. Dept. of Energy & Climate Change, London, UK.
 Manda, A.K., Heath, J.L., Klein, W.A., Griffin, M.T., Montz, B.E., 2014. Evolution of multi-well pad development and influence of well pads on environmental violations and wastewater volumes in the Marcellus shale (USA). *J. Environ. Manag.* 142, 36–45.
 Miller, S.M., Wofsy, S.C., Michalak, A.M., Kort, E.A., Andrews, A.E., Biraud, S.C., Dlugokencky, E.J., Eluszkiewicz, J., Fischer, M.L., Janssens-Maenhout, G., Miller, B.R., Miller, J.B., Montzka, S.A., Nehrkorn, T., Sweeney, C., 2013. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U. S. A.* 110 (50), 20018–20022.
 Miyazaki, B., 2009. Well integrity: an overlooked source of risk and liability for underground natural gas storage. Lessons learned from incidents in the USA. *Geol. Soc. Lond., Spec. Publ.* 313 (1), 163–172.
 Molofsky, L.J., Connor, J.A., Wylie, A.S., Wagner, T., Farhar, S.K., 2013. Evaluation of methane sources in groundwater in northeastern Pennsylvania. *Groundwater* 51 (3), 333–349.
 Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestad, J., Huang, J., Koch, G., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhan, H., 2013. Anthropogenic and Natural Radiative Forcing. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
 O'Sullivan, F., Paltsev, S., 2012. Shale gas production: potential versus actual greenhouse gas emissions. *Environ. Res. Lett.* 7, 044030.
 Olejnik, S., Algina, J., 2003. Generalized eta and omega squared statistics: measures of effect size for some common research designs. *Psychol. Methods* 8 (4), 434–447.
 Osborn, S.G., Vengosh, A., Warner, N.R., Jackson, R.B., 2011. Methane contamination of drinking water accompanying gas-well drilling and hydraulic fracturing. *Proc. Natl. Acad. Sci. U. S. A.* 108 (20), 8172–8176.
 Parker, D.E., Legg, T.P., Folland, C., 1992. A new daily Central England Temperature series 1772–1991. *Int. J. Climatol.* 12, 317–342.
 Rivard, C., Lavoie, D., Lefebvre, R., Séjourné, S., Lamontagne, C., Duchesne, M., 2014. An overview of Canadian shale gas production and environmental concerns. *Int. J. Coal Geol.* 126, 64–76.

- Sneath, R.W., Chadwick, D.R., Phillips, V.R., Pain, B.F., 1997. A UK Inventory of Methane and Nitrous Oxide Emissions From Farmed Livestock (Report to MAFF).
- Vengosh, A., Jackson, R.B., Warner, N., Darrah, T.H., Kondash, A., 2014. A critical review of the risks to water resources from unconventional shale gas development and hydraulic fracturing in the United States. *Environ. Sci. Technol.* 48, 8334–8348.
- Vidic, R.D., Brantley, S.L., Vandenbossche, J.M., Yoxtheimer, D., Abad, J.D., 2013. Impact of shale gas development on regional water quality. *Science* 340, 6134.
- Watson, T.L., Bachu, S., 2009. Evaluation of the potential for gas and CO₂ leakage along wellbores. *SPE Drill. Complet.* 24 (1), 115–126.
- Ziemkiewicz, P., Quaranta, J.D., McCawley, M., 2014. Practical measures for reducing the risk of environmental contamination in shale energy production. *Environmental Science, Processes & Impacts* 16 (7), 1692–1699.